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RECOVERY OF SILICON, COPPER AND ALUMINUM FROM SCRAP SILICON WAFERS BY LEACHING AND PRECIPITATION

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Abstract

The Silicon wafer plays an important role in modern electronic products. During the production of Silicon wafer, a large amount of scrap Silicon wafer is generated. The necessity for it to be recycled and recovered is of vast importance since the metal Silicon (Si) is the major by product. Besides Silicon, the wafers also contain a small quantity of Copper (Cu) and Aluminum (Al), that are considered as minor elements during the recovery process of the Silicon wafer production. Therefore, in order to conserve limited natural resources, minimize pollution problems and recover the valuable resource of Silicon from scrap wafer, a leaching method was adopted in this study. Physical and chemical methods such as screening and leaching processes were employed to investigate the recovery of Silicon and other metals from scrap wafers. The leaching process was carried out with three leaching reagents such as HCl, HNO_3 and H_2SO_4. Batch studies were also conducted to optimize the leaching operating conditions with consideration to the leaching time, the concentration of leaching reagent, temperature and solid/liquid ratio. The result of the leaching tests revealed that the Copper and Aluminum contained in the scrap wafers can be 100% leached and removed by using 5N HNO_3 under the conditions of two hours leaching time, 5g/50 mL of solid/liquid ratio, and 70°C temperatures. After leaching with Nitric acid, a high purity of Silicon was obtained and could be reused as the feed material for the production of Silicon wafer. The Copper and Aluminum remaining in the leaching solution was recovered through precipitation by adjusting its' pH to 11 and 7 to form $Cu(OH)_2$ and $Al(OH)_3$, respectively.

Key words: aluminum, copper, recover, scrap, silicon, wafer

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1. Introduction

The Silicon wafers are thinly, sliced semiconducting materials that are widely used in the electronics industry as a substrate for microelectronic devices and integrated circuits (IC) (van Zant, 2000). Crystalline Silicon wafers are also used in solar panels in order to catch the energy from the sun to produce electricity. Ninety percent (90%) of Silicon wafer are used in the solar industry and ten percent (10%) of Silicon wafer is used in the electronics industry. Hence, a huge amount of Silicon wafer is produced to support the development of modern electronic products. It is estimated that 250,000 Silicon wafers are used daily in the semiconductor industry for the manufacturing of electronic products (Silicon Industry, 2011).

During the production of Silicon wafers, a large amount of scrap Silicon wafer is also generated (approximately 3 million per year). According to the sage concepts forecast report, the amount of Silicon wafer production by 2015 will be 11,000 million square inches with annual growth rate of 7.3 % per year

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(http://www.sageconceptsonline.com/docs/report1.pdf).

In general, wafers are made with mono-crystalline Silicon or polycrystalline Silicon and currently, there is no substitute for these materials except the re-melting or reclamation of Silicon wafers. Besides Silicon, the wafer also contains minor amounts of Copper (Cu) and Aluminum (Al). A shortage of Silicon has been predicted from 2013 onwards, therefore, the Silicon present in the wafer is a valuable resource for recycling (Homan, 2009; Rusirawan and Farkas, 2015). In addition, both Copper and Aluminum are also valuable metals for potential industrial materials hence the necessity for recycling and recovery of these valuable metals from the scrap Silicon wafers. This is of vast importance from an economical viewpoint. It is expected that the chip manufacturers can save approximately four to six million dollars per year by recycling wafers and also reusing these wafers.

Taiwan is one of the largest suppliers of semiconductors in the world and, it occupies two-thirds of the global share in the IC manufacturing sector. According to the Polysilicon and Wafer Supply Chain Quarterly Report, the annual production of photovoltaic solar wafers in Taiwan will grow to a capacity of 30 GW by 2013 (Photovoltaic-Production, 2013). However, the directivity of the semiconductor and solar industry is also increasing simultaneously in Taiwan and it is estimated that the generation of defective or discarding Silicon wafers are also increasing correspondingly. Therefore, the need to recycle Silicon wafers is essential to maximize the environmental benefits of both the solar and semiconductor industry. In order to recover the valuable resource of Silicon from scrap Silicon wafer, this study has investigated the removal efficiency of the impurities of Copper and Aluminum by a nitric acid leaching method.

Many investigations have been reported on the recycling of photovoltaic (PV) solar panels along with Silicon wafers by thermal and chemical methods (Anja, 2006; Bohland, 1997; Doi, 2001; Eberspacher, 1997; Frisson, 2000; Fthenakis, 2000; Galan, 2005; Klugmann-Radziemska, 2010; Rusirawan and Farkas, 2015; Yamashita, 2003). However, these studies are limited to recycling of solar modules, reuse of wafers, life cycle assessment of solar modules wafers and the economic benefits of solar modules recycling. For example, Frisson et al. proposed two methods for the recycling of PV solar modules i.e., pyrolysis in a conveyer belt process and pyrolysis in a fluidized bed reactor (Frisson, 2000). However, Silicon wafers are reclaimed by following the cleaning and etching sequence of metallization with 15% HF, H_2SO_4, H_2O and HNO_3 at 80°C followed by 20% NaOH at 85°C. The energy consumption of producing new wafers and the recycling of spent wafers and reported that wafer recycling takes two hundred and fourteen (214) KW less energy than the production of a new wafer (Anja, 2006). Unfortunately, there are few reports available on the recovery of Silicon from damaged wafers. Dong et al. (2011), summarized the beneficial analysis of recycling of solar grade Silicon waste produced during preparation of solar wafer by several methods (Dong, 2011). However, these methods are either expensive in industrial scale or need to overcome negative implications in order to be practical. Research to date has shown no viable solution to recover the valuable metals Si, Cu and Al from damaged Silicon wafers.

Leaching is a widely used hydro-metallurgical technique which converts metals into soluble salts in aqueous media. In comparison with pyro-metallurgical operations, leaching is much cheaper and more flexible to be established and operated. Acid leaching is a common industrial practice for the recovery of metals from mineral ores. The most common acids used for leaching are Hydrochloric acid, Sulphuric acid and Nitric acid (Lee, 2013; Schilm, 2003). Dissolving soluble minerals from a solid ore or concentrate by Sodium hydroxide is considered as alkaline leaching (Hu et al., 2016; Pehlivan et al., 2014). Ammonium hydroxide is also used for extracting metals such as Copper and nickel from their ores (Zhao and Liuc, 2010). Mazzocchitti et al., (2009) developed a hydrometallurgical process for the selective removal of Silicon and Aluminum from the ilmenite concentrates by leaching with Sodium hydroxide solutions. Liu et al. (2014) investigate the effect of leaching temperature, leaching time, and hydrochloric acid concentration on the $ZrOCl_2$ and SiO_2 concentrations. Recently Bronsted acid ionic liquid 1-butyl-3-methyl-imidazolium hydrogen sulfate was used to leach Copper from waste printed circuit boards (Huang et al., 2014).

Precipitation is also a commonly used method to remove metal ions from solutions. The precipitation process usually adjusts pH, adds chemicals that stimulate precipitation, converting the metal ions into metal hydroxides or insoluble metal salts. Precipitation recovery of metals by chemicals or pH adjustment is the oldest method. Due to the inexpensive and simplicity of the process, precipitation is still a widely used method in recovery of precious metals from waste materials (Castro and Martins, 2009).

Several researchers have recovered Copper from electronic waste by acid leaching and precipitation process with Sodium hydroxide (Kokes et al., 2009). Kamberović et al., (2009) used bench-scale method for extraction and recovery of Copper and precious metals from waste PCBs by subjecting a serial of hydrometallurgical processing routes such as Sulphuric acid leaching and precipitation for Cu recovery. Amélie Janin et al., (2009) selectively recovered Copper from chromatids Copper arsenate by precipitation with pH adjustment.

In this study, the major composing elements of silicon wafers Silicon, Copper and Aluminum were recovered from scrap Silicon wafers by leaching technology using various leaching reagents such as HCl, HNO_3 and H_2SO_4. Further, the batch experiments were conducted to optimize leaching conditions with solid/liquid ratio, leaching time, concentration of the reagent and leaching temperature. Aluminum and Copper present in the leaching solution is recovered by precipitation and the obtained Silicon collected.

2. Experimental

2.1. Sample collection and chemicals

The Silicon wafer samples were obtained from domestic manufacturers in Taiwan. used in this study were collected from domestic manufacturers in Taiwan. Sulfuric acid (95 ~ 97%, GR reagent grade), Nitric acid (65%, GR reagent grade), and Hydrochloric acid (37%, GR reagent grade) were used as leaching reagents (purchased from Merck company). Deionized water was used for all experiments. The scrap Silicon wafers obtained from the local recyclers were already pre-grounded and screened to 50-mesh (aperture 0.297mm) size. These powder samples were of 1 - 5 g with a fixed leaching solution of 50 mL used for further experiments.

2.2. Analysis of water content, ash content, density and metal concentration

The collected silicon wafers were subjected to water content, ash content, density and metal concentration analysis. Taiwan EPA's established aqua-regia digestion method NIEA S321.63B (https://www.niea.gov.tw/) was used to analyze metal ion (Cu and Al) concentration using inductively coupled plasma-atomic emission spectrometry (ICP-AES) (Perkin Elmer, model 3300). For water content, ash content and density of ground samples, other Taiwan EPA's established industrial methods NIEA R203.02C (https://www.niea.gov.tw/) and NIEA R205.01C (https://www.niea.gov.tw/) were utilized and described in the follow paragraphs.

2.2.1. Aqua-regia digestion method

This method contains a series of the following steps: 3g of ground Silicon wafer sample was added to the 250mL round bottom flask containing 1mL H_2O for wetting the sample. To this mixture, 21mL HCl and 7mL HNO_3 solutions were added slowly and waited for 16 hours at room temperature. Then the reaction temperature was increased slowly and the solution was refluxed for 2 hours. In next step, the reaction temperature was cooled and filtered the digestion mixture. The filtrate was then diluted to a volume of 100 mL and this solution was used for the analysis of Copper and Aluminum by ICP-AES.

2.2.2. Water content

Water content present in the ground sample of silicon wafer was calculated by placing known (weight, w1) amount of sample in an oven at 105±5°C. After 2 hours, the sample was removed from the oven and kept in a desiccator. The sample weight was recorded again after attaining room temperature. This process was repeated until getting the difference less than 5mg and an average of the dried weights was calculated (w_2). The water content was calculated from the Eq. (1):

$$Water\ content(\%) = \left[\left(w_1 - w_2\right)/w_1\right] \times 100 \quad (1)$$

2.2.3. Ash content

A known weight of the ground samples (w_1) was subjected to burn in a furnace at 800±50°C for 3 hours and lower the temperature 300°C. The samples were removed from the oven and placed in a desiccator until the room temperature attained. The weight of the sample was recorded again and labelled as (w_3). The ash content was calculated as shown in the Eq. (2):

$$Ash\ content(\%) = \left(w_3/w_1\right) \times 100 \quad (2)$$

2.2.4. Combustible material

The combustible material present in ground scrap silicon wafer was calculated according to the Eq. (3):

$$Combustible\ material(\%) = 100 - \left(\begin{matrix} Water\ content\ + \\ Ash\ Content \end{matrix}\right) \quad (3)$$

2.2.5. Density

The sample density (ρ_s) was calculated by using pycometer in according to the Eq. (4).

$$\rho_s = \left(W \times \rho_w\right)/\left[w_a - \left(w_b - W\right)\right] \quad (4)$$

where ρ_w is the density of ultra-pure water, w_a = weight of ultra-pure water, W = weight of ultra-pure water, wb = weight of ultra-pure water + sample.

2.3. Leaching

Leaching process was subjected to the 50 mesh screened samples in order to separate the impurity metals Cu and Al from Si present in Silicon wafers. The operating conditions adopted in this leaching study was summarized in Table 1.

Table 1. Leaching operating conditions of scrap Silicon wafer

Leaching Reagent	*Concentration (N)*	*Temperature (○C)*	*Time (hr)*	*Solid/liquid ratio (g/mL)*
Hydrochloric acid	1, 3, 5, 10	27	1, 2, 3, 4	1g/50mL
Sulfuric acid	1, 3, 5, 10	27	1, 2, 3, 4	1g/50mL
Nitric acid	1, 3, 5, 10	27, 70	1, 2, 3, 4	1g/50mL, 3g/50mL 5g/50mL 10g/50mL

For each leaching test, 50 mL of leaching reagent was agitated with the wafer samples in a 300 mL beaker using a magnetic stirrer. The effect of the various operating parameters on leaching process such as the concentration of the leaching reagent, leaching time, solid/reagent ratio and temperature were investigated and optimized by batch method.

The content of metal from its leaching solution of each leaching test was calculated as follows (Eq. 5):

$$Leaching\ recovery\ (\%) = \left(\frac{w_l}{w_t}\right) \times 100 \quad (5)$$

where: w_l = weight of metal leached, w_t = weight of metal contained in testing sample

Copper metal dissolves readily in dilute acids to form Cu (II) ions along with hydrogen gas. Similarly, Aluminum forms Al (III) ions with the liberation of hydrogen gas. The chemical reactions involved in the leaching of the metals (Copper and Aluminum) with acid reagents were as follows (Eqs. 6-7):

$$Cu_{(s)} + H^+X^-{}_{(aq)} \rightarrow Cu^{2+}{}_{(aq)} + H_{2(g)} + X^-{}_{(aq)} \quad (6)$$

$$Al_{(s)} + H^+X^-{}_{(aq)} \rightarrow Al^{3+}{}_{(aq)} + H_{2(g)} + X^-{}_{(aq)} \quad (7)$$

2.4. Precipitation

After separation of the Silicon as a solid in the leaching process, the optimal leaching solution contained the metals, Cu and Al. These metals from the leaching solution were recovered by the precipitation process via pH adjustments. The pH was adjusted with Sodium Hydroxide. In order to investigate the effect of pH on precipitation of Al and Cu, the pH value of the optimal leaching solution was adjusted to 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11 and 12. The efficiency of pH precipitation of various metals under different pH was calculated as follows (Eq. 8):

$$Precipitation\ efficiency\ (\%) = \frac{(w_i - w_f)}{w_i} \times 100 \quad (8)$$

where: w_i is the weight of metal in leaching solution before any pH adjustment and w_f is the weight of metal in leaching solution after pH adjustment.

2.5. SEM-EDS analysis

To investigate the weight percentage and appearance of Al and Cu product, SEM-EDS studies were carried out with multi-function scanning electron microscope (JSM-6400) at required magnification. The Scanning Electron Microscope was equipped with an Energy Dispersive Spectrometer (EDS). The samples were dried at a temperature of 105°C and deposited on a brass holder under vacuum. The beam accelerating voltage was maintained 15 kV with the secondary electron image as a detector.

3. Results and discussion

3.1. Analysis of water content, ash content, density and metal concentration

The collected scrap Silicon wafer powder had a size of less than 50 mesh (<0.297 mm). Metals Cu and Al present in scrap silicon wafer samples were analyzed by ICP-AES and the average contents were found to be 0.14 and 0.13 weight percentage respectively. The contents of water, ash, combustible material and density of the ground samples were 0.78%, 104.81%, 0% and 2.67g/cm^3, respectively. Thus, most of the scrap Silicon wafers was composed of inorganic materials.

3.2. Leaching

Leaching experiments were conducted in batch process with desired particle size (<0.297 mm) of scrap wafers. Optimization of leaching conditions was tested with various leaching reagents, leaching temperature, time of leaching and solid/liquid ratio. Since Silicon is the major composing element in the scrap wafers, in order to obtain the high purity Silicon, the leaching experiments were focused on the removal and recovery of Al and Cu.

3.2.1. Effect of time on leaching

To investigate the effect of leaching time on the removal of Al and Cu from scrap Silicon wafers, the experiments were conducted at different times of 1, 2, 3 and 4 hours. 50 mL of 5N of each leaching reagent (HCl, HNO_3 and H_2SO_4) was added to 1g of wafer sample and agitated at 27°C. From Fig. 1, it was observed that the leaching efficiency increases with increasing the contact time of the metal with the reagent. However, the Aluminum removal efficiency was higher than Copper, as Aluminum leaching efficiency was 100% with all leaching reagents. Leaching efficiencies of Al in HCl were 100% from 1 hr onwards; these efficiencies were at least two-fold higher to that of Cu. The Cu leaching increased from about 24% to 53% as the reaction time increased from one to four hours. The leaching efficiency of Al and Cu with H_2SO_4 was lower than the HCl as Al removal was increased from 75% to 100% with increasing time from one to four hours, but Cu removal was increased 5% to 19% with the same leaching time, which was at least five times lower than the Al removal.

With HNO_3 as the leaching reagent, the removal efficiency of Al and Cu was higher than the other two before mentioned leaching reagents and achieved 100% for both metals. Among all the leaching reagents, Nitric acid acted as the most efficient reagent in the removal of both Al and Cu. Therefore, the optimum leaching time could be stated as two hours. The result of leaching efficiencies of these metals with leaching time was depicted in Fig. 1

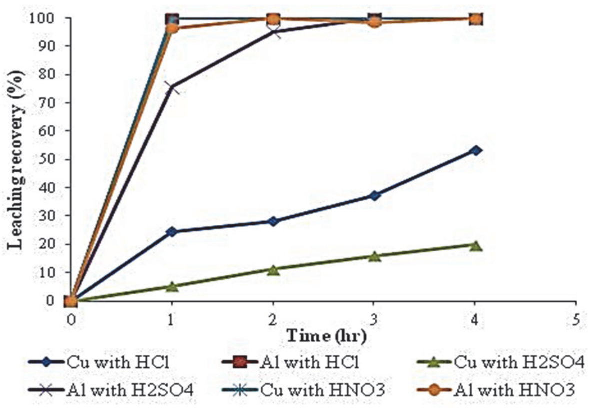


Fig. 1. Effect of time on leaching efficiency of Cu and Al with various leaching reagents (Concentration of leaching reagent = 5N; solid/liquid ratio = 1g/50m and Temperature = 27°C)

3.2.2. Effect of reagent concentration on leaching

The effect of leaching reagent concentration on the removal of Al and Cu was studied by varying the regent concentration from 1N to 10N at the conditions of 2 hr leaching time, 1g/50mL solid/liquid ratio and 27°C temperature. The results were presented in Fig. 2. From the Fig. 2, it was observed that the leaching efficiency (%) in the removal of Al generally increased with an increase in concentration of HCl leaching reagent, however, with H_2SO_4 leaching reagents the removal efficiency was increased from 1N to 3N and then decreased. The removal of Cu was lower than Al for HCl and H_2SO_4 at all concentrations. However, the leaching efficiency of Cu and Al with Nitric acid increased from 1N to 5N and thereafter decreasing. Albeit, the trend was increasing from 1N to 5N, but 100% removal was achieved at 5N concentration of HNO_3 for both Cu and Al.

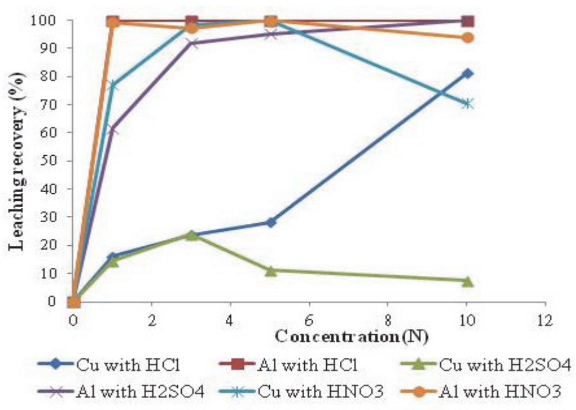


Fig. 2. Effect of concentration of reagent on leaching efficiency of Cu and Al (leaching time = 2h; solid/liquid ratio = 1g/50mL and Temperature = 27°C)

Therefore, Nitric acid proved to be the best dissolving agent for metals in comparison to both Hydrochloric and Sulfuric acid, as these agents may give rise to the issues with precipitation. More so, due to the low cost, regeneration and reuse of HNO_3, this acid has proven to be the best leaching reagent in metallurgical process.

3.2.3. Effect of temperature and solid/liquid ratio on leaching

The effect of quantity on scrap Silicon wafer powder in 50 mL of leaching solution on the recovery efficiency of Copper and Aluminum was studied by using the solid/liquid ratio 0.02 (1g/50mL), 0.06 (3g/50mL), 0.1 (5g/50mL) and 0.2 (10g/50mL) with 5N concentration of HNO_3 solution, two hours contact time at 27°C and 70°C. The impregnation of different solid/liquid ratios with a recovery rate was presented in Fig. 3.

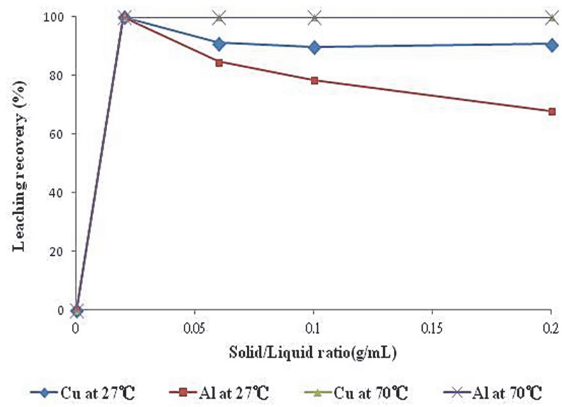


Fig. 3. Effect of solid/liquid ratio and temperature on leaching efficiency of Cu and Aln(leaching time = 2h; and Concentration of leaching reagent HNO3 = 5N

From the figure, it was observed that as the solid/liquid ratio was increasing from 0.02 to 0.2, the leaching recovery of Al and Cu present in scrap Silicon wafer was decreasing at 27°C for HNO_3 solution. 100% recovery of both the metals was achieved with 0.02 solid/liquid ratio at the above mentioned conditions and then decreased to 89% of Copper and 61% of Aluminum with increasing solid/liquid ratio up to 0.2 percent. The results also indicate that the recovery of Al and Cu present in scrap Silicon wafer powder was very rapid by the leaching acid. However, the leaching efficiency of Cu and Al was obtained 100% with increasing temperature to 70°C for all solid/liquid ratios. Although leaching efficiency was maximum for all solid/liquid ratio of wafer/HNO_3 at the specified conditions and also with the use of high solid/liquid ratio, it proved to be more economical, except for the filtration of the solution with high solid/liquid ratio (0.2 g/mL) presenting some difficulty. Therefore, in order to recover maximum amounts of Cu and Al, this study was chosen with the optimum temperature and solid/liquid ratio condition of 70°C and 5g/50mL (0.1). Thus, the optimum leaching conditions finalized in this investigation were HNO_3 as a leaching reagent with 5N concentration, 2 hr leaching time, 5g/50mL solid/liquid ratio and temperature 70°C. At these conditions, Silicon present in the scrap wafer samples were obtained as a solid in the leaching solution, and further separated by filtration.

3.3. Precipitation

The pH test results with HNO_3 leaching reagent was illustrated in Fig. 4. Precipitation method was employed to separate and recover Cu and Al present in the obtained optimal leaching solution by varying pH of the leaching solution. Since HNO_3 was used as the leaching reagents, the obtained optimal leaching solution had a very low pH. The metal dissolved in the solution can be converted into metal precipitate by changing the solution's pH value. Thus, the pH value of this optimal leaching solution was adjusted by the addition of sodium hydroxide and HNO_3.

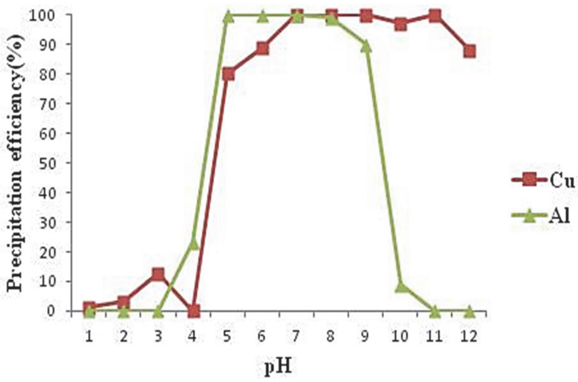


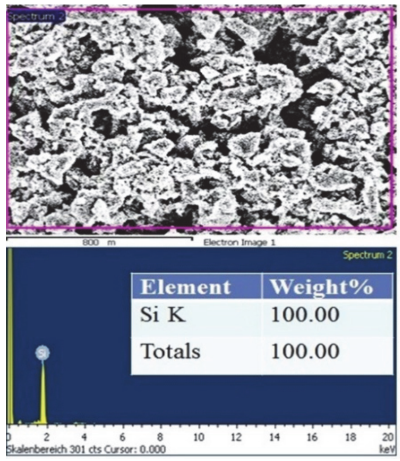
Fig. 4. Effect of pH on precipitation recovery of Cu and Al from optimum leaching solution

The pH values investigated in these tests range from 1 ~ 12. At the end of each pH adjustment, the metal concentrations of Cu and Al in the solution were determined by ICP-AES. Then the pH adjustment efficiency of each metal was calculated according to the Eq. (4). Copper can be removed by precipitation as $Cu(OH)_2$ at pH ranging from 9 to 11. Fig. 4 illustrates the metal Cu could be successfully separated from Al under the investigated pH that Cu began to precipitate at pH>1 and Al precipitation from pH 4 to pH 11. The speciation of Al was dependent upon the pH of the solution. Due to the amphoteric nature of Al, it changed to anionic form under strong alkaline conditions at pH >8. The dominant form of Al at acidic pH range was Al^{+3}. At low pH, inorganic acids readily complex with dissolved Aluminum, and can increase its equilibrium solubility in solution. The precipitation recovery of Cu increased from 13% to 100% with increasing pH value from 1 to 7, and follows the same up to pH 11. Al precipitates 23% at pH 4 and reaches 100% sharply when increasing pH to 5, and maintains the same recovery of 100% until pH 8.

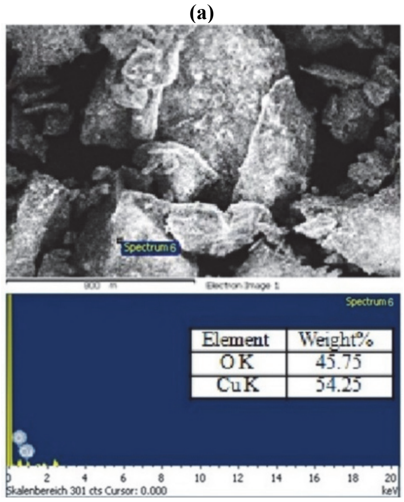
The Al contained in the solution can be separated from Cu by adjusting the solution's pH value to 11, as Cu precipitates completely at that pH. Then the Al contained solution can be precipitation at pH 7. Therefore, precipitation with pH adjustment method was applied successfully as a best option for the separation and complete recovery of Cu and Al with maximum purity.

3.4. SEM-EDS analysis

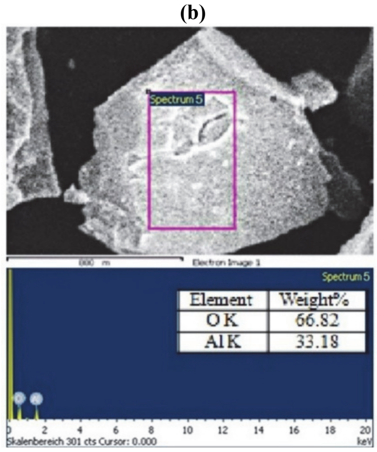
After the recovery of Si, Al and Cu from Nitric acid leaching solution, these products from scrap Silicon wafer were analyzed by SEM-EDS (Fig. 5) to investigate the purity of metals.



(a)



(b)



(c)

Fig. 5. SEM-EDS analysis of obtained Silicon, Copper hydroxide and Aluminum: (a) Silicon; (b) Copper hydroxide, (c) Aluminum

Energy dispersion spectra confirms the purity of Silicon 100% as expected and also noticed that there were no further contaminated or interfering compounds present in this product. The weight percent of Cu and Al in the precipitate were 54 and 33 which was reasonably good compared to its theoretical weight percent of Cu (65%) and Al (35%) in Cu(OH)2 and Al(OH)3, respectively.

4. Conclusions

Silicon wafer waste from the local solar semiconductor industry was subjected to the treatment process comprising leaching and precipitation. The major composing element of scrap Silicon wafer was Si and minor metals Cu, and Al were successfully recycled and recovered.

The results showed a promising outcome and led to propose a conceptual Silicon recovery strategy from Silicon wafer waste. Batch studies were conducted with leaching time, concentration of the leaching reagent, solid/liquid ratio and temperature to optimize leaching conditions. The optimal leaching conditions such as leaching reagent, concentration of leaching reagent, solid/liquid ratio and temperature drawn from the experiments were HNO_3 with 5N concentration, 2 hr, 5g/50mL, and 70°C, respectively.

Under these conditions, Cu and Al were recovered 100% by leaching. Precipitation method with pH adjustment was applied to Nitric acid leaching solution for the separation and recovery of metals Cu and Al.

The Cu recovery achieved in this process was 100% at pH 11 as a product of $Cu(OH)_2$ and the Al recovery achieved in this process was 100% at pH 7 as a product of $Al(OH)_3$. Therefore, this study concludes that leaching and precipitation method with pH adjustment could be used to recover the valuable elements of Si, Cu and Al from mix scrap Silicon wafer in order to reach the goals of resource recycling.

Acknowledgments

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